



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : Custodero et al.
Serial No. : 09/583,655 Examiner: Boss, W.
Filed : May 30, 2000 Group Art: 1775
For : CARBON BLACK COATED WITH AN ALUMINOUS LAYER
AND PROCESS FOR OBTAINING SAME

#9
KW
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DECLARATION OF ARNAUD LAPRA
UNDER 37 C.F.R. § 1.132

EXPRESS MAIL NO: ET 346771091 US

RECEIVED
DEC 05 2002
TC 1700

Assistant Commissioner for Patents
Washington, D.C. 20231

I, Arnaud LAPRA residing at 2, rue des Bouleaux, 63100 Clermont-Ferrand, FRANCE, do declare:

1. I currently hold the position of searcher at Michelin & CIE in the chemistry research department, where I have been employed since September 1999. I have been working more particularly in the field of rubber compositions and their additives under the supervision of E.CUSTODERO, a co-inventor of the invention described and claimed in the above-identified application. I have been informed of and understand said invention.

2. The instant invention relates to a composition, specifically a novel, modified carbon black which has (1) at least a partial coat of aluminum oxide and/or aluminum

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hydroxide; (2) a BET surface area of 30-400 m²/g; (3) an average particle size of 20-400 nm; and (4) an ultrasound disagglomeration rate greater 1x10⁻³ mm⁻¹/s.

3. Silica-modification of carbon black is not equivalent to carbon black modified with aluminum oxide and/or hydroxide.

4. I have conducted tests to measure and compare the physical properties of a conventional reinforcing filler of untreated carbon black N234 (#1 control), an alumina and/or aluminum hydroxide surface-treated carbon black of the invention (#2), and a conventional, silica surface-treated carbon black (#3). The results are presented in the following tables, which demonstrate the properties of three compositions before curing (Table A) and after curing (Table B) at 150°C for 40 minutes. The amounts of the different products are expressed in phr, *i.e.*, parts per 100 parts by weight of elastomer.

TABLE A

COMPOSITION	#1 untreated	#2 Al-treated	#3 silica-treated
SBR ¹	100	100	100
N234	50	--	--
Modified N234	--	50	--
CRX 2000 ²	--	--	50
Si69	--	5	5
ZnO	3	3	3
Stearic acid	1.5	1.5	1.5
Antioxidant ³	1	1	1
DPG ⁴	0.5	0.5	0.5

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6. The results presented in the above tables are expressed in comparative values, with the value "100" assigned to the reference Composition #1, which contains untreated carbon black. The Mooney plasticity and the moduli were determined in accordance with the methods taught in the instant specification.

7. The "Bound Rubber" test allows one to determine, in a composition that has not been vulcanized, the proportion of elastomer that is intimately associated with the reinforcing filler, and thus insoluble in typical organic solvents. This value provides an objective and quantitative indication of the reinforcing activity of the filler in the rubber composition. The "Bound Rubber" test is a well-recognized method for characterizing the reinforcement quality provided to the rubber composition by the filler. The method has been described, for example, in Plastics, Rubber and Composites Processing and Applications, Vol. 25, No. 7, p. 327 (1996), and in Rubber Chemistry and Technology, Vol. 69, p. 325 (1996). The "Bound Rubber" test has been specifically used to determine the amount of elastomer bound to carbon black (*see, e.g.*, French Standard NF T 45-114 (June 1989)).

8. In the "Bound Rubber" tests conducted to generate the data in Table B, the amount of remaining elastomer (*i.e.*, unextractable with toluene) was measured after treating 300-350 mg of the test rubber composition with 80-100 ml of toluene for fifteen days in the dark at room temperature, followed by vacuum drying at 100°C for 24 hours. The amount of "bound rubber" (% by weight) was calculated from the difference between initial and final weights, taking into account components initially present that are naturally insoluble.

9. As illustrated in Table B, the Mooney plasticity value was found to be significantly lower for the sample containing the alumina and/or aluminum hydroxide-coated
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Sulfur	1.5	1.5	1.5
CBS ⁵	2	2	2

¹ butadiene-styrene copolymer² silica-modified carbon black sold by the company CABOT³ N-1,3-dimethylbutyl-N-phenylparaphenylenediamine⁴ Diphenylguanidine⁵ N-cyclohexyl-2-benzothiazylsulphenamide**TABLE B**

COMPOSITION	#1 untreated	#2 Al-treated	#3 silica-treated
Plasticity (MU) ¹	100	95	125
M10 (Mpa)	100	98	115
M100 (Mpa)	100	107	124
M300 (Mpa)	100	101	128
"Bound Rubber" (%)	100	133	110

¹ Mooney Plasticity Values expressed in Mooney Units ("MU").

5. The composition containing conventional, silica surface-treated carbon black, which is represented as Composition #3 in the above tables, was prepared following the teachings of Section III-2 at pages 24-25 of the instant specification. The formulations of Composition #1 (having untreated carbon black) and Composition #2 (having alumina and/or aluminum hydroxide surface-treated carbon black) are identical to that presented in Table 2 at page 29 of the instant specification. The formulation of Composition #3 differs from Composition #2 only in that the filler is a silica surface-treated black sold under reference CRX 2000 by the company CABOT.

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carbon black of the invention (#2) compared to the control sample containing untreated carbon black (#1), which reflects the excellent ability of composition #2 to be worked in the uncured state. *See also* Section III-2 of the instant specification.

10. In direct contrast, the sample containing the silica-treated carbon black (#3) demonstrated significantly inferior workability in the uncured state, with a Mooney Plasticity Value which was greater than 25% higher compared to the control sample containing untreated carbon black (#1 in Table B). The compromised workability exhibited by compositions containing the silica-treated carbon black is unacceptable for industrial applications.

11. Therefore, the increase of moduli values (M10, M100, M300) presented in Table B is actually indicative of the increased rigidity of the composition, which reflects the higher Mooney plasticity value, and not a result of any reinforcing effect.

12. The "Bound Rubber" values illustrated in Table B confirm the strong interaction between the rubber and filler, which is significantly stronger in the sample containing the alumina and/or aluminum hydroxide-coated carbon black of the invention (#2) than in the sample containing silica-treated carbon black (#3), or in the control sample containing untreated carbon black (#1). Specifically, the "Bound Rubber" value of the sample containing the alumina and/or aluminum hydroxide-coated carbon black of the invention (#2) is 21% greater than that of the sample containing silica-treated carbon black (#3), and 33% greater than that of the control sample containing untreated carbon black (#1).

13. The high "Bound Rubber" value of the composition containing an

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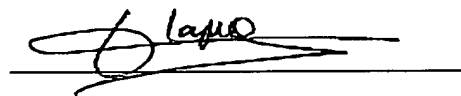
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alumina and/or aluminum hydroxide-coated carbon black of the invention represents a very significant increase in the reinforcing properties of the filler. This enhanced reinforcing property is an unexpected and surprising result when viewed by a person of ordinary skill in the art.

14. An important technical advantage is gained by incorporating a composition containing an alumina and/or aluminum hydroxide-coated carbon black of the invention into tires, which results in tires with significantly better wear resistance.

15. I hereby declare that all statements made herein by my own knowledge are true, and that all statements made on information and belief are believed to be true, and further that I make these statements with the knowledge that willful false statements, and the like, are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of any patent issuing from the above-identified application.

Dated: 26/11/2002



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BAKER BOTTS L.L.P.



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Sir:

Transmitted herewith is:

- [X] An Amendment in response to the May 30, 2002 Office Action for the above-identified patent application (17 pages).
- [X] A Declaration Under 37 C.F.R. § 1.132.
- [X] Fee Calculation.
- [X] Check in the amount of \$956.00 in payment of the three month extension of time, and excess claim fee.
- [X] In the event that an additional extension of time is required in connection with this submission, including an extension of time under 37 C.F.R. § 1.136, applicants request such an extension and authorize the Commissioner to charge payment of any extension of time fee to Deposit Account No. 02-4337. Duplicate copies of this sheet are enclosed.
- [X] The Commissioner is hereby authorized to charge payment of any additional filing fees required, including and fees due under 37 C.F.R. § 1.16 and/or 37 C.F.R. § 1.17 or to credit any overpayment to Deposit Account No. 02-4337. Duplicate copies of this sheet are enclosed.
- [X] An Extension of Time to respond to the PTO communication dated May 30, 2002 is hereby requested. The required fee is enclosed herewith.

The Fee has been calculated as shown below:

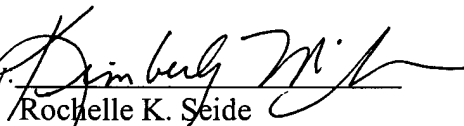
<u>FOR</u>	(Col. 1) <u>No. Filed</u>		(Col. 2) <u>No. Extra</u>		Small Entity <u>Rate</u>	<u>Fee</u>	OR	Other Than A <u>Small Entity</u> <u>Rate</u>	<u>Fee</u>
Total Claims	24	- 22 =	2	x	9 =	\$0.00	x	18 =	\$36.00
Ind. Claims	3	- 3 =	0	x	42 =	\$0.00	x	84 =	\$0.00
Multiple Dependent Claim				+	140 =		+	280 =	\$0.00
Excess Claim Fee Total						\$			<u>\$36.00</u>

Extension for response (check only one):

	<u>SMALL ENTITY</u>	<u>OTHER THAN A SMALL ENTITY</u>
Within first month	<input type="checkbox"/> \$ 55	<input type="checkbox"/> \$ 110
Within second month	<input type="checkbox"/> 200	<input type="checkbox"/> 400
Within third month	<input type="checkbox"/> 460	<input checked="" type="checkbox"/> 920
Within fourth month	<input type="checkbox"/> 720	<input type="checkbox"/> 1,440
<input type="checkbox"/> Other:		
<input checked="" type="checkbox"/> Total Fee Due: <u>\$ 956.00.</u>		

BAKER BOTTS L.L.P.

Dated: December 2, 2002

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